Frustrated quantum-spin system on a triangle coupled with e_g lattice vibrations - Correspondence to Longuet-Higgins *et al.*'s Jahn-Teller model -

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We investigate the quantum three spin model $(\mathbf{S_1}, \mathbf{S_2}, \mathbf{S_3})$ of spin= 1/2 on a triangle, in which spins are coupled with lattice-vibrational modes through the exchange interaction depending on distances between spin sites. The present model corresponds to the dynamic Jahn-Teller system $E_g \otimes e_g$ proposed by Longuet-Higgins *et al.*, Proc.R.Soc.A.**244**,1(1958). This correspondence is revealed by using the transformation to Nakamura-Bishop's bases proposed in Phys.Rev.Lett.**54**,861(1985). Furthermore, we elucidate the relationship between the behavior of a chiral order parameter $\hat{\chi} = \mathbf{S_1} \cdot (\mathbf{S_2} \times \mathbf{S_3})$ and that of the electronic orbital angular momentum $\hat{\ell}_z$ in $E_g \otimes e_g$ vibronic model: The regular oscillatory behavior of the expectation value $\langle \hat{\ell}_z \rangle$ for vibronic structures with increasing energy can also be found in that of $\langle \hat{\chi} \rangle$. The increase of the additional anharmonicity(chaoticity) is found to yield a rapidly decaying irregular oscillation of $\langle \hat{\chi} \rangle$.

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The triangular Heisenberg antiferromagnets play an important role in our understanding the resonating valence bond(RVB) state, in which the scalar chirality for three spins $\mathbf{S_1} \cdot (\mathbf{S_2} \times \mathbf{S_3})$ is expected to have a nonzero expectation value[1, 2, 3, 4]. This subject has been a focus of recent experimental activities[5, 6, 7], since it was expected that a frustrated $s = 1/2$ triangular antiferromagnet lattice might be realized in $NaTiO_2$ and $LiNiO_2$[8].

In this Letter, we propose a triangular cluster model of the Heisenberg antiferromagnet in which quantum spins are coupled with lattice vibrations, for the purpose to discuss magnetic properties in relation to the typical dynamical Jahn-Teller system $E_g \otimes e_g$. In short, the spin-lattice interaction is introduced by expanding the exchange interaction with respect to deviation of lattice displacements from equilibrium. We shall address to the following issue: With use of a unitary transformation for this spin system, the proposed model becomes equivalent to that of the well-known vibronic problem for $E_g \otimes e_g$ Jahn-Teller system[9].

Let us consider the quantum spin system where three spins of spin=1/2 are localized at lattice sites 1,2 and 3 on triangle. The coupling between neighboring spins are expressed by the antiferromagnetic exchange interactions J_A, J_B and J_C as shown in Fig.1. The corresponding Heisenberg Hamiltonian is

$$\mathcal{H} = J_A \mathbf{S_1} \cdot \mathbf{S_2} + J_B \mathbf{S_2} \cdot \mathbf{S_3} + J_C \mathbf{S_3} \cdot \mathbf{S_1}. \qquad (1)$$

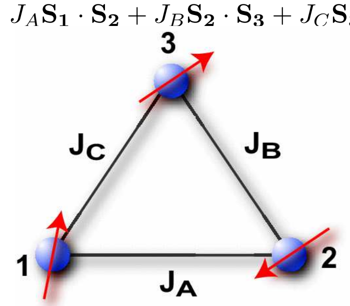

FIG. 1: Triangle with antiferromagnetic spins.

We concentrate our attention on the spin state where z component of the total spin satisfies $s_{1z}+s_{2z}+s_{3z} = 1/2$. Therefore, these bases are expressed explicitly as $|\downarrow\uparrow\uparrow\rangle, |\uparrow\downarrow\uparrow\rangle, |\uparrow\uparrow\downarrow\rangle$, where arrows denote s_{jz} for site j. By using these bases, we obtain the exchange Hamiltonian,

$$\mathcal{H}/\left(-\frac{\hbar^2}{4}\right) = \begin{array}{c} \\ \langle\downarrow\uparrow\uparrow| \\ \langle\uparrow\downarrow\uparrow| \\ \langle\uparrow\uparrow\downarrow| \end{array} \begin{array}{c} \begin{array}{ccc} |\downarrow\uparrow\uparrow\rangle & |\uparrow\downarrow\uparrow\rangle & |\uparrow\uparrow\downarrow\rangle \end{array} \\ \begin{pmatrix} -J_A + J_B - J_C & 2J_A & 2J_C \\ 2J_A & -J_A - J_B + J_C & 2J_B \\ 2J_C & 2J_B & J_A - J_B - J_C \end{pmatrix} \end{array}. \qquad (2)$$

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Next we introduce the interaction between the spins and lattice vibrations, noting the dependence of J_A, J_B and J_C on the distances between spin sites. As for the lattice vibration, we employ the normal modes for the triangle; The normal e_g modes Q_1 and Q_2, which are degenerate, are given in Fig.2. The remaining a_{1g} mode(:the

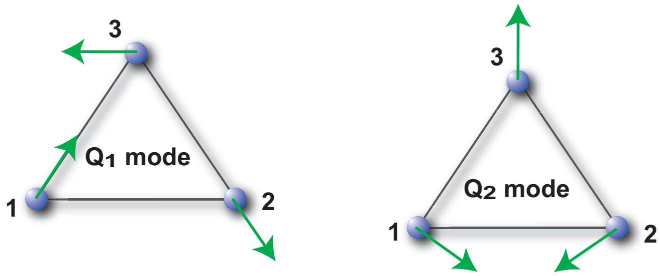

FIG. 2: The normal modes Q_1 and Q_2 in the triangle.

breathing mode) has a much higher strain energy and is ignored hereafter. (There are other global degrees of freedom related to translation of the center of mass and to rotation around the axis perpendicular to the triangular plane. They however have nothing to do with lattice vibrations and are also ignored.) Then the spin-lattice interaction is obtained as a result of the the expansion of J_A, J_B and J_C linearly in the e_g modes as follows:

$$\begin{aligned} J_A &= J\cdot\left[1+\frac{\alpha}{2}(Q_1-\sqrt{3}Q_2)\right] \\ J_B &= J\cdot[1-\alpha Q_1] \\ J_C &= J\cdot\left[1+\frac{\alpha}{2}(Q_1+\sqrt{3}Q_2)\right], \end{aligned} \tag{3}$$

where α is the coupling constant.

Concerning the spin system, on the other hand, we introduce the following bases introduced by Nakamura and Bishop for the triangular spin plaquet[10, 11, 12]:

$$\begin{aligned} |k=0\rangle &= \frac{1}{\sqrt{3}}(|\downarrow\uparrow\uparrow\rangle+|\uparrow\downarrow\uparrow\rangle+|\uparrow\uparrow\downarrow\rangle) \\ |k=\frac{2\pi}{3}\rangle &= \frac{1}{\sqrt{3}}\left(|\downarrow\uparrow\uparrow\rangle+e^{\frac{2\pi}{3}i}|\uparrow\downarrow\uparrow\rangle+e^{-\frac{2\pi}{3}i}|\uparrow\uparrow\downarrow\rangle\right) \\ |k=-\frac{2\pi}{3}\rangle &= \frac{1}{\sqrt{3}}\left(|\downarrow\uparrow\uparrow\rangle+e^{-\frac{2\pi}{3}i}|\uparrow\downarrow\uparrow\rangle+e^{\frac{2\pi}{3}i}|\uparrow\uparrow\downarrow\rangle\right). \end{aligned} \tag{4}$$

These bases reflect clockwise and anticlockwise rotations of a spin configuration on the plane of the triangle. The wave numbers $k=0,\pm 2\pi/3$ correspond to phase factors in Bloch's theorem for the system with the discrete rotational symmetry. From the viewpoint of the ligand-field theory[13], the construction of these bases (4) from $|\downarrow\uparrow\uparrow\rangle, |\uparrow\downarrow\uparrow\rangle$ and $|\uparrow\uparrow\downarrow\rangle$ is regarded as a formation of E_g and A representations in D_{3d} symmetry from the triply-degenerate T_{2g} ones in O_h symmetry. By using this new bases, the Hamiltonian matrix (2) can be transformed to

$$\mathcal{H}/\left(-\frac{3}{4}\hbar^2 J\right) = \begin{array}{c} \\ \langle k=0| \\ \langle k=\frac{2\pi}{3}| \\ \langle k=-\frac{2\pi}{3}| \end{array} \begin{array}{c} \begin{array}{ccc} |k=0\rangle & |k=\frac{2\pi}{3}\rangle & |k=-\frac{2\pi}{3}\rangle \end{array} \\ \begin{pmatrix} 1 & 0 & 0 \\ 0 & -1 & \alpha(-Q_1-iQ_2) \\ 0 & \alpha(-Q_1+iQ_2) & -1 \end{pmatrix} \end{array}. \tag{5}$$

From Eq.(5) we find that the $k=0$ manifold is completely separated from other manifolds, i.e., $\mathcal{H}=\mathcal{H}_{k=0}\otimes\mathcal{H}_{k=\pm 2\pi/3}$. $\mathcal{H}_{k=0}$ and $\mathcal{H}_{\pm 2\pi/3}$ correspond to A and E_g representation, respectively. The interaction Hamiltonian $\mathcal{H}_{k=\pm 2\pi/3}$ can result in a pair of adiabatic energy surfaces, which together with the harmonic term ($\propto Q_1^2+Q_2^2$), forms the Mexican hat potential. In fact, by applying the unitary transformation:

$$U=-\frac{1}{\sqrt{2}}\begin{pmatrix} 1 & i \\ -1 & i \end{pmatrix}, \tag{6}$$

we can get

$$\begin{aligned} \tilde{\mathcal{H}}_{k=\pm 2\pi/3} &= U^{-1}\mathcal{H}_{k=\pm 2\pi/3}U \\ &= \frac{3}{4}\hbar^2 J\mathbf{I}-\frac{3\alpha}{4}\hbar^2 J\begin{pmatrix} Q_1 & +Q_2 \\ +Q_2 & -Q_1 \end{pmatrix}. \end{aligned} \tag{7}$$

This expression accords with the electron-lattice interaction part of the vibronic Hamiltonian for the Jahn-Teller system $E_g \otimes e_g$,

$$\mathcal{H}_{JT} = \frac{1}{2}\omega^2(Q_1^2 + Q_2^2) + \alpha' \begin{pmatrix} Q_1 & +Q_2 \\ +Q_2 & -Q_1 \end{pmatrix}. \quad (8)$$

Thus, we would like to emphasize that the present system for quantum spins on the triangle coupled with doubly-degenerate vibrational e_g modes is equivalent to the vibronic model for the $E_g \otimes e_g$ system that is extensively and intensively investigated in the context of the dynamical Jahn-Teller problem.

Before proceeding to the argument on the chiral order parameter of the spin system, we shall recall the definition of the electronic orbital angular momentum in the dynamical Jahn-Teller system. For the Hamiltonian consisting of the kinetic energy $(1/2(P_1^2 + P_2^2))$ and $\mathcal{H}_{JT}$ of (8), the p-th eigenstate of the $\ell = 1/2$ manifold, $\Psi_{p,1/2}$ is given by

$$\Psi_{p,1/2} = a_{1,p}\psi_{1,0}\phi_+ + a_{2,p}\psi_{2,1}\phi_- + a_{3,p}\psi_{3,0}\phi_+ + a_{4,p}\psi_{4,1}\phi_- + \ldots \quad (9)$$

where the $\psi_{n,m}$'s are the eigenfunctions of the isotropic two-dimensional harmonic oscillator (n and m are radial and azimuthal quantum numbers, respectively), and ϕ_+ and ϕ_- are degenerate electronic states $\phi_\pm = d_u \pm i d_v$. The expansion (9) was found by rewriting $\mathcal{H}_{JT}$ in (8) into a suitable form with the use of $\phi_\pm$[14]. In the context of the spin-lattice system under consideration, the block matrix $\mathcal{H}_{k=\pm 2\pi/3}$ in (5) already takes such a suitable form with the use of Nakamura-Bishop's bases $|k = \pm 2\pi/3\rangle$, and the vibronic wave function takes the same form as (9).

In the vibronic state $\Psi_{p,1/2}$ in the dynamical Jahn-Teller system, the expectation value of the electronic orbital angular momentum $\hat{\ell}_z$ is given as

$$\begin{aligned}\langle \hat{\ell}_z \rangle_p &= \langle \Psi_{p,1/2} | \hat{\ell}_z | \Psi_{p,1/2} \rangle \\ &= \sum_{n=1}^{\infty} |a_{n,p}|^2 (-1)^{n-1} \Xi_\ell \qquad (p = 1, 2, \ldots). \end{aligned} \quad (10)$$

Here, Ξ_ℓ is the expectation value of $\hat{\ell}_z$ in the electronic states ϕ_+ and ϕ_-:

$$\Xi_\ell = \langle \phi_+ | \hat{\ell}_z | \phi_+ \rangle = -\langle \phi_- | \hat{\ell}_z | \phi_- \rangle. \quad (11)$$

The emergence of an outstanding regular oscillation of $\langle \hat{\ell}_z \rangle_p$ as a function of energy(p) was pointed out three decades ago[15], which has received a renewed attention recently in the context of nonlinear dynamics[14].

Now let's come back to the argument of the characteristic operator for the quantum spin system. With use of the bases (4), we evaluate the expectation values for chiral order parameter

$$\hat{\chi} = \mathbf{S_1} \cdot (\mathbf{S_2} \times \mathbf{S_3}). \quad (12)$$

The order parameter $\hat{\chi}$ characterizes degree of the frustration of the triangular antiferromagnet[4]. The expectation values of $\hat{\chi}$ in each of the E_g states (4) are

$$\begin{aligned}\langle k = \frac{2\pi}{3} | \hat{\chi} | k = \frac{2\pi}{3} \rangle &= -\frac{\sqrt{3}}{4} \equiv -\Xi_\chi \\ \langle k = -\frac{2\pi}{3} | \hat{\chi} | k = -\frac{2\pi}{3} \rangle &= \frac{\sqrt{3}}{4} \equiv \Xi_\chi. \end{aligned} \quad (13)$$

(The value $\langle k = 0 | \hat{\chi} | k = 0 \rangle = 0$ is now irrelevant since $|k = 0\rangle$ is coupled only with the higher frequency a_{1g} mode.) Thus, the states $|k = \pm\frac{2\pi}{3}\rangle$ and chiral order parameter $\hat{\chi}$ in the spin-lattice system correspond to states $|\phi_\pm\rangle$ and $\hat{\ell}_z$ in the dynamical Jahn-Teller system, respectively. Taking the eigenstates similar to (9), the dependence of $\langle \hat{\chi} \rangle_p$ on the p-th eigenstate is given by

$$\langle \hat{\chi} \rangle_p = \sum_{n=1}^{\infty} |a_{n,p}|^2 (-1)^n \Xi_\chi. \quad (14)$$

This means that the behavior of $\langle \hat{\chi} \rangle_p$ can be revealed by applying the analysis of $\langle \hat{\ell}_z \rangle_p$ in (10). In fact, the expectation values $\langle \hat{\chi} \rangle_p$ in (14) shows regular oscillation with increasing the energy(see Fig.3(a)), just as in the case of $\langle \hat{\ell}_z \rangle_p$ in the dynamical Jahn-Teller system[15].

Finally we note a role of the anharmonic term involved in the triangular three particle system. Let us introduce Toda-lattice potential[16]

$$U(x) = \frac{c}{d} e^{-dx} + cx - \frac{c}{d}, \quad (15)$$

where x is the deviation of inter-particle distance from the equilibrium lattice constant. c and d are constant with the condition of $cd > 0$. The total lattice potential is a sum of $U(x)$ with x the three kind of deviations for three segments of the regular triangle. In the limit $d << 1$ under the constraint $cd =$ constant, we obtain the following expansion in x:

$$\begin{aligned} U(x) &= \frac{c}{d}(1 - dx + \frac{d^2}{2!}x^2 - \frac{d^3}{3!}x^3 + \ldots) + cx - \frac{c}{d} \\ &= \frac{cd}{2}x^2 - \frac{cd^2}{6}x^3 + \ldots . \end{aligned} \quad (16)$$

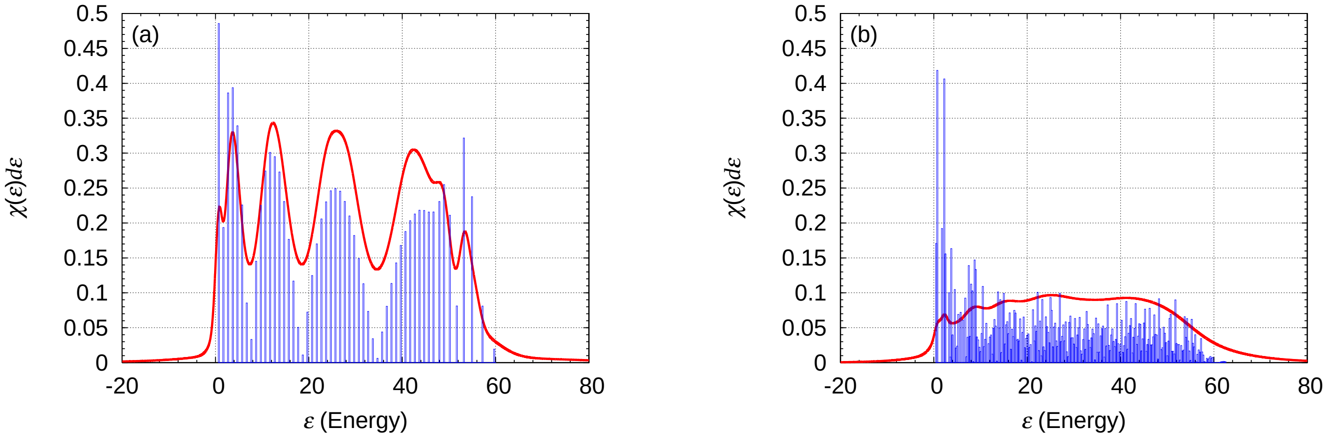

FIG. 3: Energy(ε) dependence of partially-averaged chirality $\chi(\varepsilon)d\varepsilon(=\sum_p' |\sum_{n=1}^{\infty}(-1)^n a_{n,p}^2|d\varepsilon)$ with $\varepsilon = 0.25$ in unit of Ξ_χ. (a) and (b) correspond to $\gamma = 0, 1.41$, respectively. $\alpha = 0.707$ and the unit of energy is $\hbar\omega$. Envelop functions are also constructed by Gaussian coarse-graining of each peak. (Reproduced from [14] and interpreted in the context of a triangular spin model.)

Suppressing a high-frequency a_{1g} mode and noting the symmetry of the e_g modes in Fig.2, the bilinear term in (16) leads to the 2-d harmonic oscillator potential. On the other hand, the cubic term in (16) leads to the trigonal(anharmonic) potential

$$V_A = V_A(Q_1, Q_2) = -\frac{\gamma}{3}(Q_1^3 - 3Q_1Q_2^2) \qquad (17)$$

with $\gamma = cd^2/2$ in terms of normal e_g modes Q_1 and Q_2. The chaotic semiclassical dynamics induced by the anharmonic term was explored in the context of the dynamical Jahn-Teller system[14], and we can expect a rapidly decaying irregular oscillation of $\langle\hat{\chi}\rangle$ by increasing the anharmonicity(chaoticity)(see Fig.3(b)).

In conclusion the frustrated quantum spin system on a triangle coupled with lattice vibrations is equivalent to $E_g \otimes e_g$ Jahn-Teller system. The chiral order parameter $\hat{\chi}$ should signify a quantum chaos induced by the coupling between quantum spins and lattice vibrations, and the energy dependence of $\langle\hat{\chi}\rangle$ shows the transition from regular to irregular oscillations by adding the anharmonicity.

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